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PHOTOCHEMICAL IGNITION STUDIES I. LASER IGNITION OF FLOWING PREMIXED GASES

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February 1985

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ABERDEEN PROVING GROUND, MARYLAND

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Gas mixtures of CH_4/air, CH_4/N_2O, C_3H_8/air, C_3H_8/N_2O, and C_2H_2/air were ignited above a slot burner by focusing three different laser beams whose wavelengths are 193 nm, 248 nm, and 532 nm. Minimum ignition energies were determined over a wide range of equivalence ratios and over the energy range of 0.1-40 mjoule/pulse for each gas/laser combination. A substantial wavelength dependence of the minimum ignition energy was observed for the ArF (193 nm) and KrF (248 nm) lasers acting on the different gas mixtures and was

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attributed to molecule-specific multiphoton-induced photochemistry of the various fuel and oxidizer molecules. Here, the necessary radicals and/or ions which are needed to cause ignition to occur are apparently produced in a controlled way. The Nd:YAG second harmonic (532 nm) laser exhibited a much smaller minimum ignition energy range for the various gas mixtures due to a different ignition mechanism involving gas breakdown, i.e., a laser-produced spark. This process appears to be much harder to control with respect to energy deposition than the photochemical one. The most efficient laser-driven ignition system was the one where the ArF (193 nm) laser acted on C_2H_2/air and laser energies as low as 0.2 mjoule caused ignition. Our results further indicate that there should be a number of ways to improve the efficiency of the photochemical ignition process. Thus, this new type of ignition source appears to possess considerable potential for utility in both practical applications as well as in allowing direct, time-resolved studies of the chemistry of ignition itself, which is an area of considerable current interest.

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I. INTRODUCTION

The study of ignition is of fundamental interest to all research concerned with initiation of the heat releasing pathways of energetic materials. For the Army, there is obviously great interest in assuring that weapons performance will not suffer by problems, such as delayed ignition. Proper ignition is particularly difficult for the newly developed low vulnerability propellants (LOVA's), as well as for new charge designs in which the projectile intrudes sufficiently into the propellant bed so that the available length for the primer tube is shortened. Many of these ignition problems can be approached and solved empirically by systematic alterations and subsequent testing of the particular charge design, but there still exists a great need to understand the fundamental aspects of the ignition process itself in which chemistry plays an important role. In fact, even though the general area of ignition has been under study for a very long time, the details of the chemistry involved are generally unknown, except for some of the simplest gas phase reactive mixtures.

There has been much phenomenological work done on electric spark ignition of gaseous mixtures.[1] In these studies, a closed bomb was utilized where different gaseous mixtures were introduced at different pressures and ignited by an electric spark generated between two electrodes whose distance from each other could be varied. In this manner, a family of curves were produced in which the minimum ignition energy was plotted against the percent fuel at different total pressures. As alternative ignition sources, lasers have certain unique characteristics. They can be propagated long distances, even to remote and inhospitable areas, potentially without much loss of available energy. This energy can be imparted on a very short time scale, i.e., in the 10 nsec regime for the ArF, KrF, and Nd:YAG lasers used here, as opposed to the microsecond regime for spark ignition. In fact, lasers have been studied for quite some time for their potential as ignition sources for premixed gases [2,3,4] as well as for propellant ignition.[5]

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[1] B. Lewis and G. Von Elbe, "Combustion, Flames and Explosions of Gases," Academic Press, New York, p. 390, 1951.

[2] J.H. Lee and R. Knystausas, "Laser Spark Ignition of Chemically Reactive Gases," AIAA Journal, Vol. 7, p. 312, 1969.

[3] F.J. Weinberg and J.R. Wilson, "A Preliminary Investigation of the Use of Focussed Laser Beams for Minimum Ignition Energy Studies," Proc. Roy. Soc., Lond. A, Vol. 321, p. 41, 1971.

[4] R.W. Schmieder, "Laser Spark Ignition and Extinction of a Methane-Air Diffusion Flame," J. Appl. Phys., Vol. 52, p. 3000, 1981.

[5] P.P. Ostrowski, J.F. Grant, J. Sharma, W.L. Garrett, D.S. Downs, and S. Krasner, "Laser Ignition of Double and Triple-Based Gun Propellants," 17th JANNAF Combustion Meeting, Langley, VA, Vol. II, p. 175, September 1980.

One of the major difficulties that was found in laser ignition was that the process was generally quite uncontrollable since it involved gas breakdown, i.e., the production of a laser-produced spark, which was usually accompanied by a substantial blast wave. This blast wave was typically intense enough to cause transition into detonation for detonable gas mixtures at less than one atmosphere pressure[2,3] or to cause extinction of a atmospheric pressure flame already stabilized on a laboratory burner.[4]

Another characteristic unique to lasers operating in the ultraviolet, is that if their wavelength is short enough, then they can be used to induce photochemistry. Thus, instead of just heating the sample and causing thermal chemistry to occur at the associated temperature or causing a relatively uncontrolled laser-produced spark, the energy is now used to break photochemically accessible bonds and to create specific intermediate species with a given energy distribution which depends on the photophysical details of the process. The possibility, therefore, exists that laser energy can be coupled very efficiently and in a controlled manner into desired photochemical channels.

The utility of photochemical ignition was recognized some years ago where initially the work involved flash photolysis,[6] a xenon arc lamp[7] and later an excimer laser operating on the F_2 (157 nm) and ArF (193 nm) lines.[8] In this latter experiment, H_2/O_2 and H_2/air mixtures were irradiated and the results were analyzed on the basis of a single-photon photolysis model where O atom laser production from O_2 was the initial step, followed by subsequent secondary reactions leading to full combustion. More recently, a substantial effort to understand the chemical details of ignition has been undertaken in which a flashlamp single-photon photolysis ignition source is being used in conjunction with a molecular beam mass spectrometer sampling apparatus.[9]

Very recent flame studies, where short wavelength lasers were utilized for the detection of flame atoms, particularly O and H, have demonstrated that multiphoton photolysis of fuel and oxidizer molecules can occur at room temperature, as well as in a combustion environment, depending on the presence of appropriate photochemical precursors as well as sufficient laser pulse

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[6]R.G.W. Norrish, "The Study of Combustion by Photochemical Methods," Tenth Symposium (International) on Combustion, The Combustion Institute, p. 1, 1965.

[7]A.E. Cerkanowicz and J.G. Stevens, "Case Studies of the Simulation of Novel Combustion Techniques," Proceedings of 1979 Summer Computer Simulation Conference, p. 170, July 1979.

[8]M. Lavid and J.G. Stevens, "Photochemical Ignition of Premixed Hydrogen/Oxidizer Mixtures with Excimer Lasers," Technical Meeting of Eastern Section/The Combustion Institute, Providence, RI, 1983.

[9]R. Peterson, D. Lucas, F.C. Hurlbut, and A.K. Oppenheim, "Molecular Beam Overrun in Sampling Transient Combustion Processes," J. Phys. Chem., Vol. 88, pp. 4746-4749, 1984.

energy.[10,11] A subsequent study has probed some of the details of the multiphoton photochemical interaction between the ArF (193 nm) laser and a number of simple hydrocarbons.[12] All of this recent work from our laboratory has provided us with the impetus and some element of insight into the subject area covered in this report, i.e., multiphoton-induced photochemical ignition. For this report, five different gas mixtures were ignited by three different lasers focused over a slot burner. Two different initiation mechanisms must be invoked to explain our results. The first is attributed to multiphoton photochemistry at 193 nm and 248 nm and the second to the onset of gas breakdown at 532 nm resulting in a laser-produced spark.

II. EXPERIMENTAL

The experimental apparatus is quite simple and is illustrated in Figure 1. The ArF (193 nm) and KrF (248 nm) lasers result from mixing appropriate gas mixtures in a commercial excimer laser, a Lumonics Model 861M. Typically, laser energies up to 12 mJ/pulse (ArF) and 35 mJ/pulse (KrF) are employed at a pulse rate of 10 pps. The rectangular laser beams are masked down to a smaller beam profile whose dimension is ~2.2 x 1.5 cm^2 and are subsequently focused by a nominally 100 mm focal length lens to a point approximately 3 mm above the slot of a slot burner whose orifice dimensions are 0.5 mm x 60 mm. The second harmonic beam of a Quanta-Ray Nd:YAG laser (532 nm) is similarly focused as that of the excimer lasers and its pulse energy varied by changing the lamp energy output of the oscillator. The pulse energy for the excimer lasers is varied by introducing different partially transmitting filters (Acton Research) into the beam path.

The gases are taken from standard grade cylinders without further purification. The oxidizer and fuel gases are passed through Matheson #602 Rotameters, with a maximum flow rate ~10^3 scc/min. Typically, the oxidizer flow was fixed, while the fuel flows were varied. The laser energies were measured by volume absorbing calorimeters (Scientech), which also acted as beam stops (see Figure 1).

It was observed that neither of the two excimer laser beams could cause air breakdown, i.e., a laser-produced spark, to occur up to their maximum output energies. This is probably due to the fact that the highly divergent beams would not yield a sufficiently tight focus and thus the power density at the focal point was not high enough. The 532 nm beam, however, caused air breakdown to occur at about the 10 mJ/pulse level.

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[10] A.W. Miziolek and M.A. DeWilde, "Multiphoton Photochemical and Collisional Effects During Oxygen-Atom Flame Detection," Optics Letters, Vol. 9, p. 390, 1984.

[11] A.W. Miziolek and M.A. DeWilde, "Photochemical and Collisional Aspects of Laser Diagnostics of Combustion," Army Science Conference, West Point, 1984.

[12] A.W. Miziolek, R.C. Sausa, and A.J. Alfano, "Efficient Detection of Carbon Atoms Produced by Argon Fluoride Laser Irradiation of Simple Hydrocarbons," ARBRL-TR-in preparation.

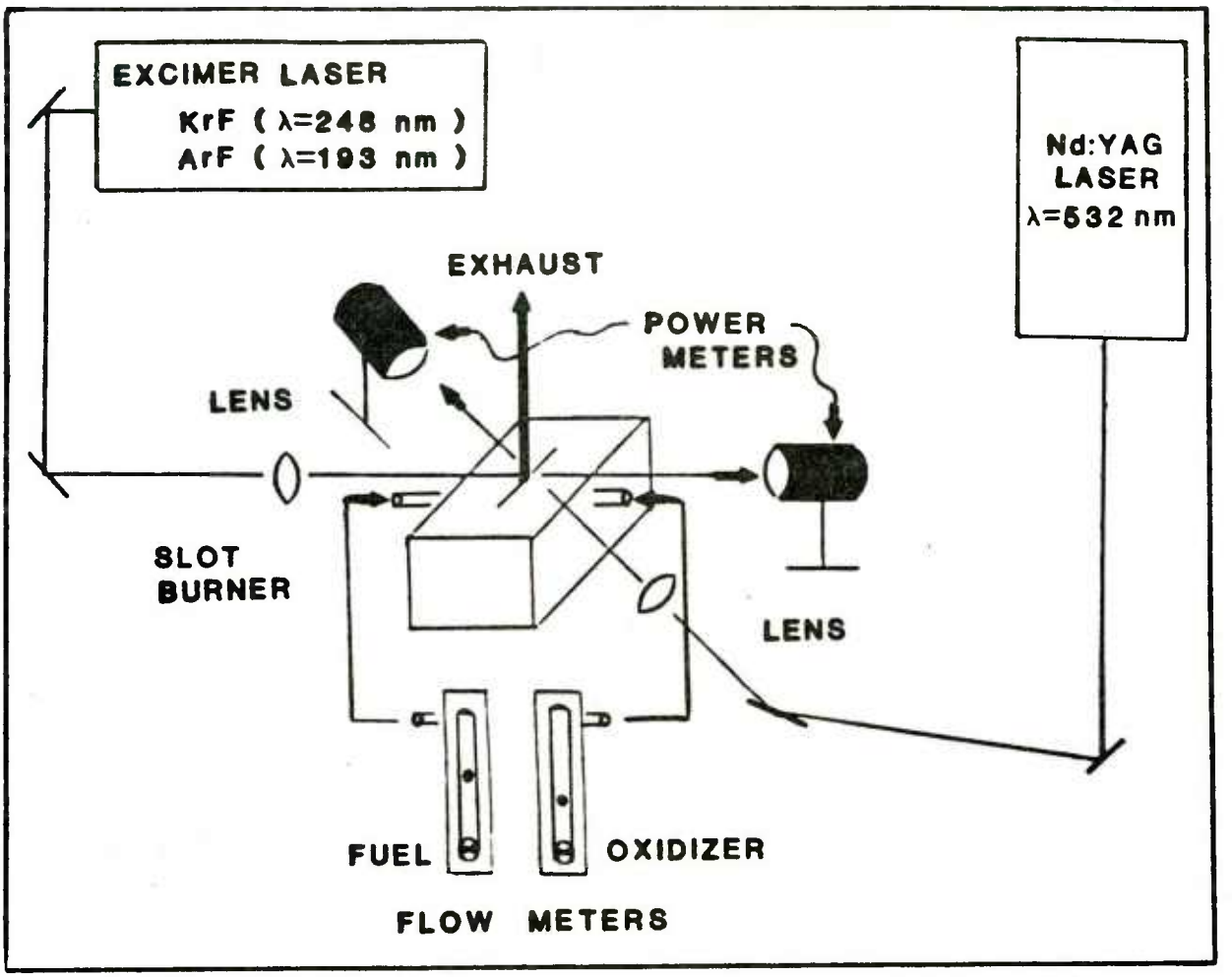

Figure 1. Experimental Schematic

The operational criterion for ignition was the appearance of a flame on the burner within the first three laser shots (this corresponded to approximately the time it took to open and close the laser trigger switch). It was found, for example, that a gas mixture just at the threshold of ignition might ignite after a number of laser shots. This, however, was not surprising since typically the pulse-to-pulse laser amplitude stability is $\sim \pm$ 10%.

All of the laser energies were measured before flowing the premixed gases through the burner and thus any problems in energy measurements due to absorption and/or scatter of laser radiation by the premixed gases were avoided. It was found that, for the excimer laser ignition case, most of the available laser energy was transmitted with relatively little (25% or less) being absorbed or scattered both below the ignition energy threshold and once ignition occurred. For the Nd:YAG second harmonic (532 nm) case, however, once the gas breakdown threshold was exceeded, then most of the laser energy appeared to be absorbed and/or scattered.

III. RESULTS AND DISCUSSION

The dependence of minimum ignition energy on percent fuel for CH_4/air is given in Figure 2. For this particular gas mixture, neither the ArF nor KrF excimer laser could initiate ignition up to their maximum output energies. The Nd:YAG laser did, however, ignite the mixture and, for comparison, a similar plot for the spark ignition case in the closed bomb is included.[1] The graph for CH_4/N_2O is given in Figure 3. In this case, the ArF laser was found to ignite the mixture very efficiently, whereas the KrF laser had no effect. An explanation for this behavior might include the possibility that the ArF laser couples photochemically well with the N_2O molecule, whereas, the KrF does not. Previous experiments have shown that multiphoton-induced photochemistry of N_2O occurs at 225.6 nm in which O atoms are produced.[10,11]

The graph for C_3H_8/air is given in Figure 4. Here the ArF laser was found to ignite the mixture, but the KrF laser did not. The fact that the ArF laser was able to ignite the C_3H_8/air mixture appears to be consistent with our observations that the ArF laser is photochemically more active with C_3H_8 than with CH_4.[12] This assumes, however, that the observed low pressure behavior extrapolates to atmospheric pressure. For C_3H_8/N_2O mixtures, the data is given in Figure 5. In this case, all three lasers were found to ignite the gases, but with quite different efficiencies. It is of interest to note that both excimer lasers exhibit curve shapes similar to those found for electric spark ignition while the Nd:YAG has a "flat" dependence up to the upper limit of rotameter flow rates, a behavior exhibited for all gas mixtures studied.

The gas mixture most active with laser ignition is C_2H_2/air and the corresponding graph is shown in Figure 6. For comparison, we also include the graph for C_3H_8/air mixtures (we could not find similar data for C_2H_2/air) spark ignited in a closed bomb.[1] Two observations are worth noting. One, for the case of ArF laser ignition, the minimum ignition energy is very similar to that found for spark ignition, i.e., 0.2-0.25 mJ. Two, the shape of the KrF and ArF curves is surprisingly flat in the full-rich equivalence ratio range. This could be due to the presence of possible experimental artifact that may affect all of our results presented in Figures 2-6 to some extent.

The nature of this artifact is air entrainment, since, for reasons of experimental simplicity, the premixed gases were flowed directly into laboratory air and thus, as compared to a closed bomb experiment, a certain amount of mixing with this air may be occuring at 3mm above the burner slot. The extent of mixing would depend on the individual gas mixtures being studied. The presence of this artifact is supported by the fact that many gas mixtures seem to ignite and burn outside the range of flammability on the fuel-rich side. The ignition outside the range of flammability, which is mostly observed for the Nd:YAG case, is probably due to the relatively sizable laser-produced spark which encompasses regions where laboratory air has mixed in with the premixed gases thus resulting in local equivalence ratios within the flammability range. Similarly, for the case of C_2H_2/air ignited by the excimer lasers, (Figure 6) beyond the fuel-rich flammability region, ignition causing radicals and/or ions are probably produced in sufficient numbers within a large enough volume in which regions of air entrainment are found where the local equivalence ratio is again within the flammability limit. In

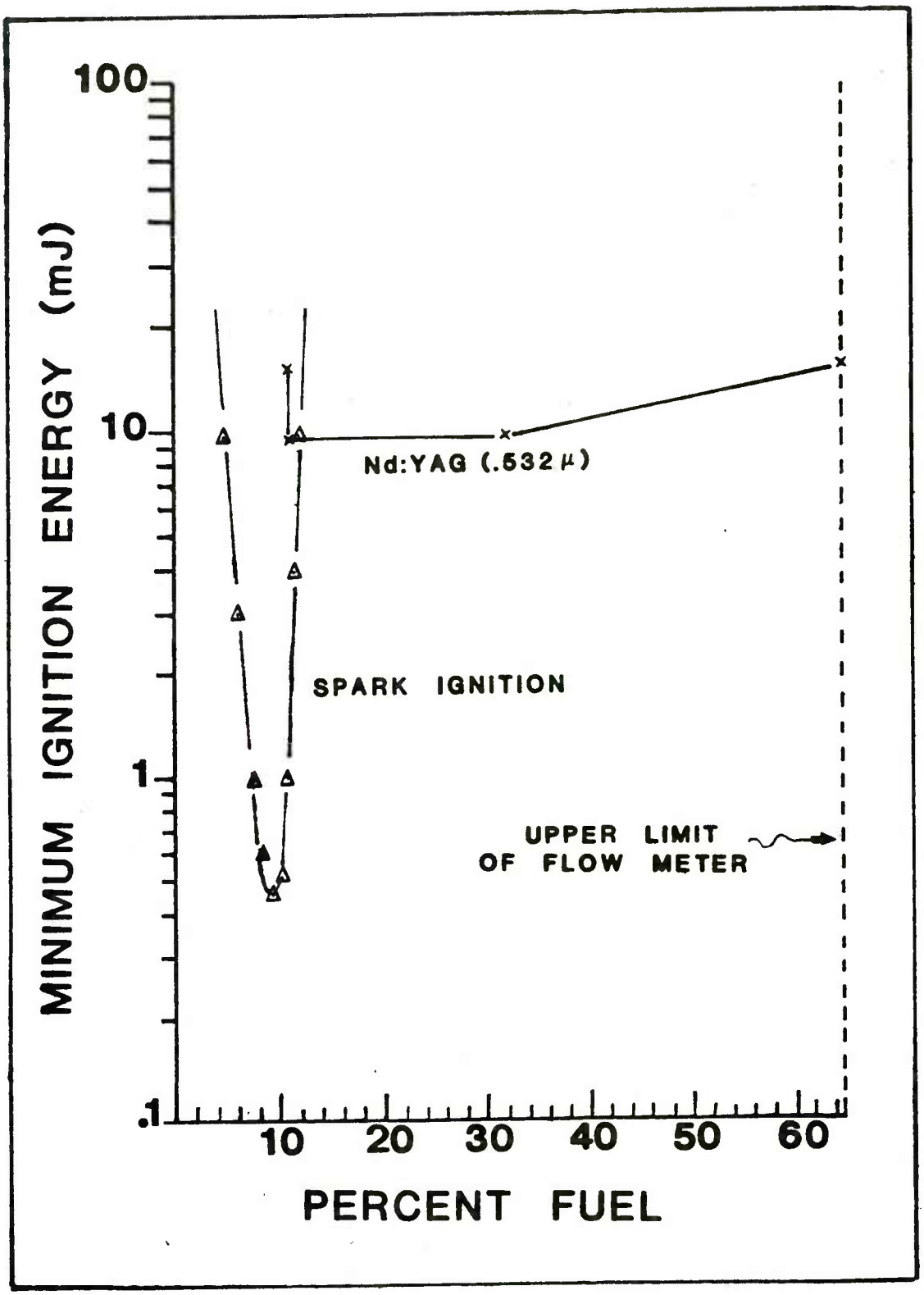

Figure 2. Dependence Of Minimum Ignition Energy On Percent Fuel For CH_4/Air. A Plot For CH_4/Air Spark Ignition In A Closed Bomb Is Included For Comparison.

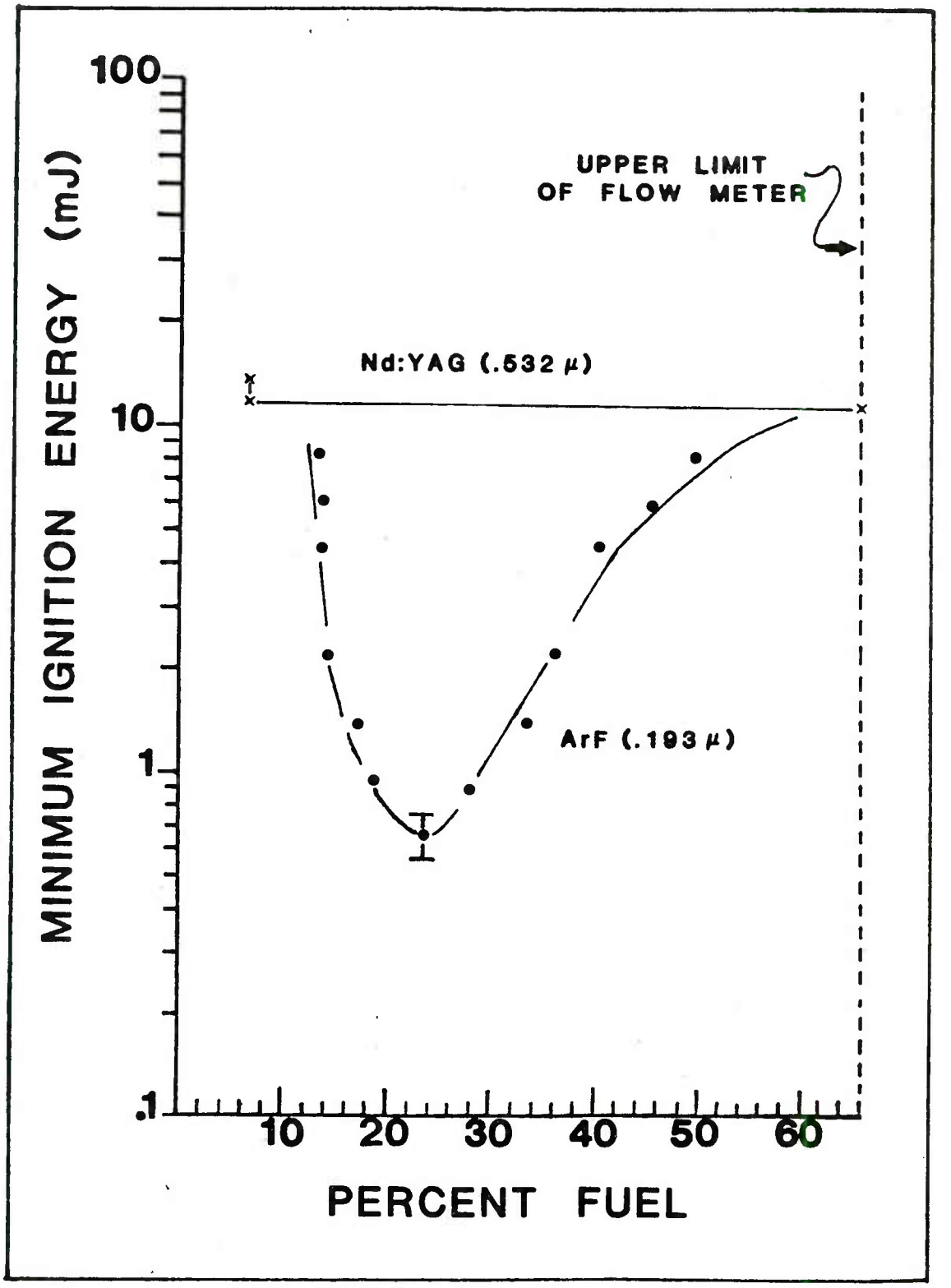

Figure 3. Dependence Of Minimum Ignition Energy On Percent Fuel For CH_4/N_2O.

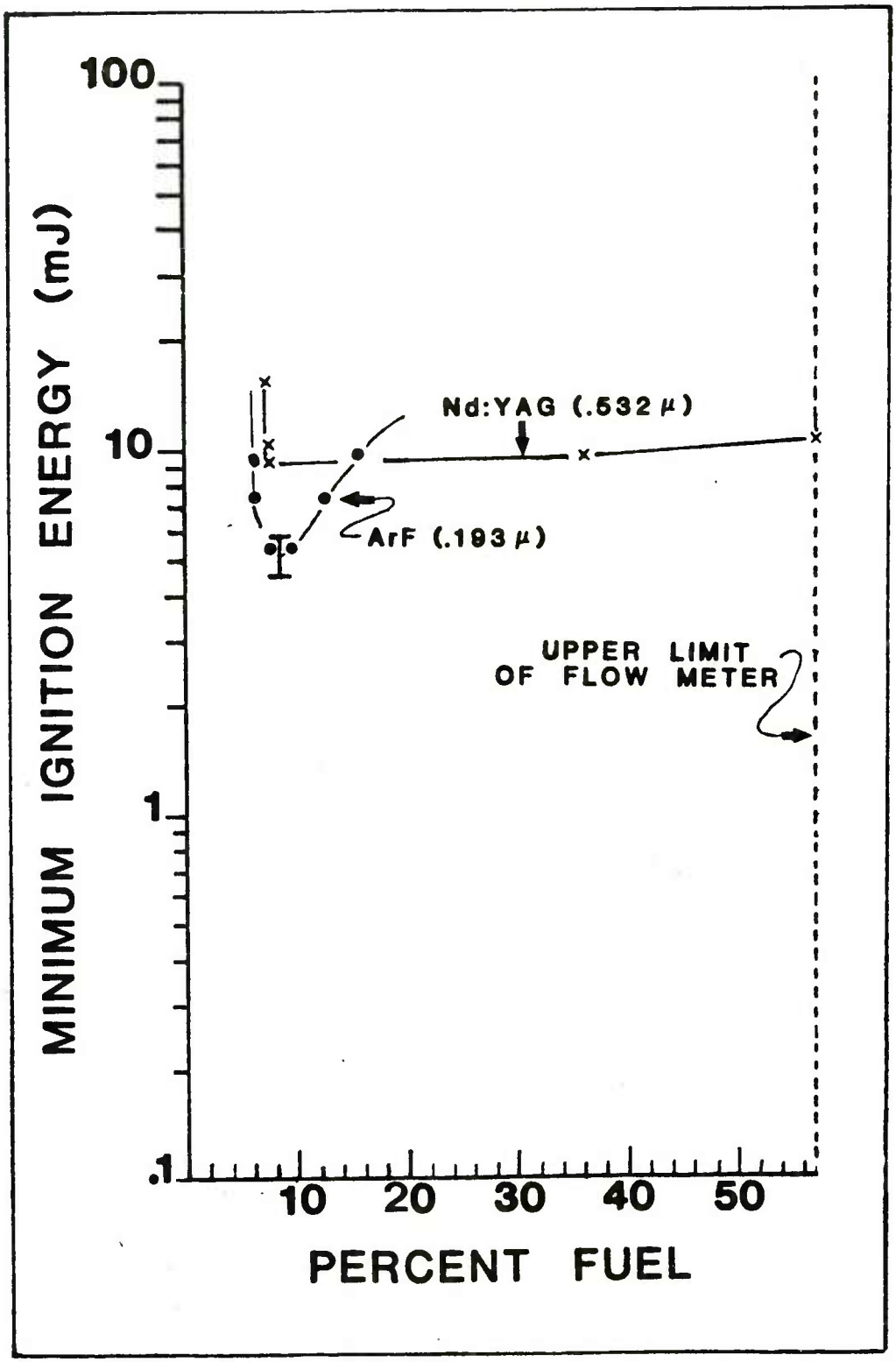

Figure 4. Dependence Of Minimum Ignition Energy On Percent Fuel For C_3H_8/Air

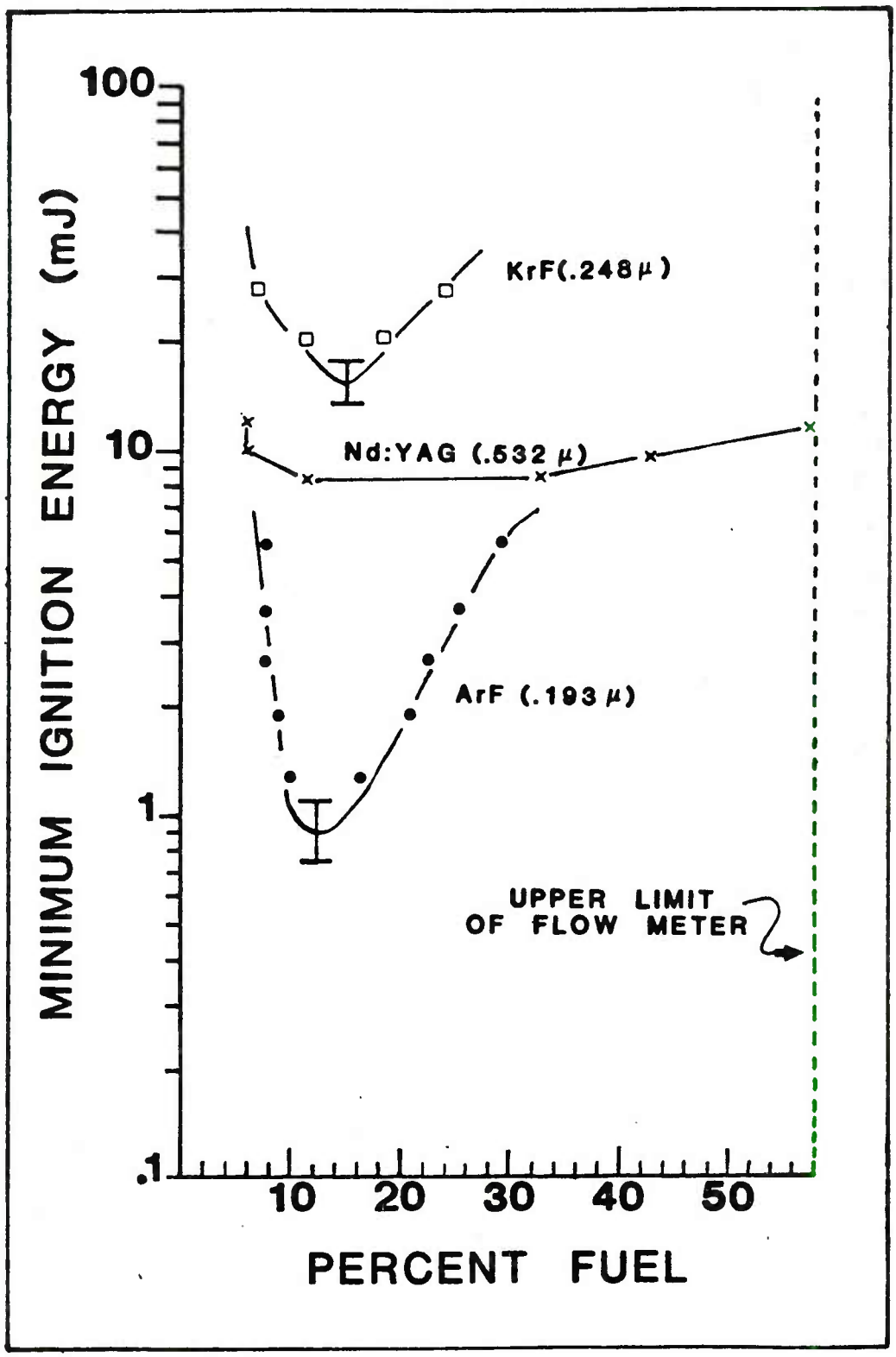

Figure 5. Dependence Of Minimum Ignition Energy On Percent Fuel For C_3H_8/N_2O.

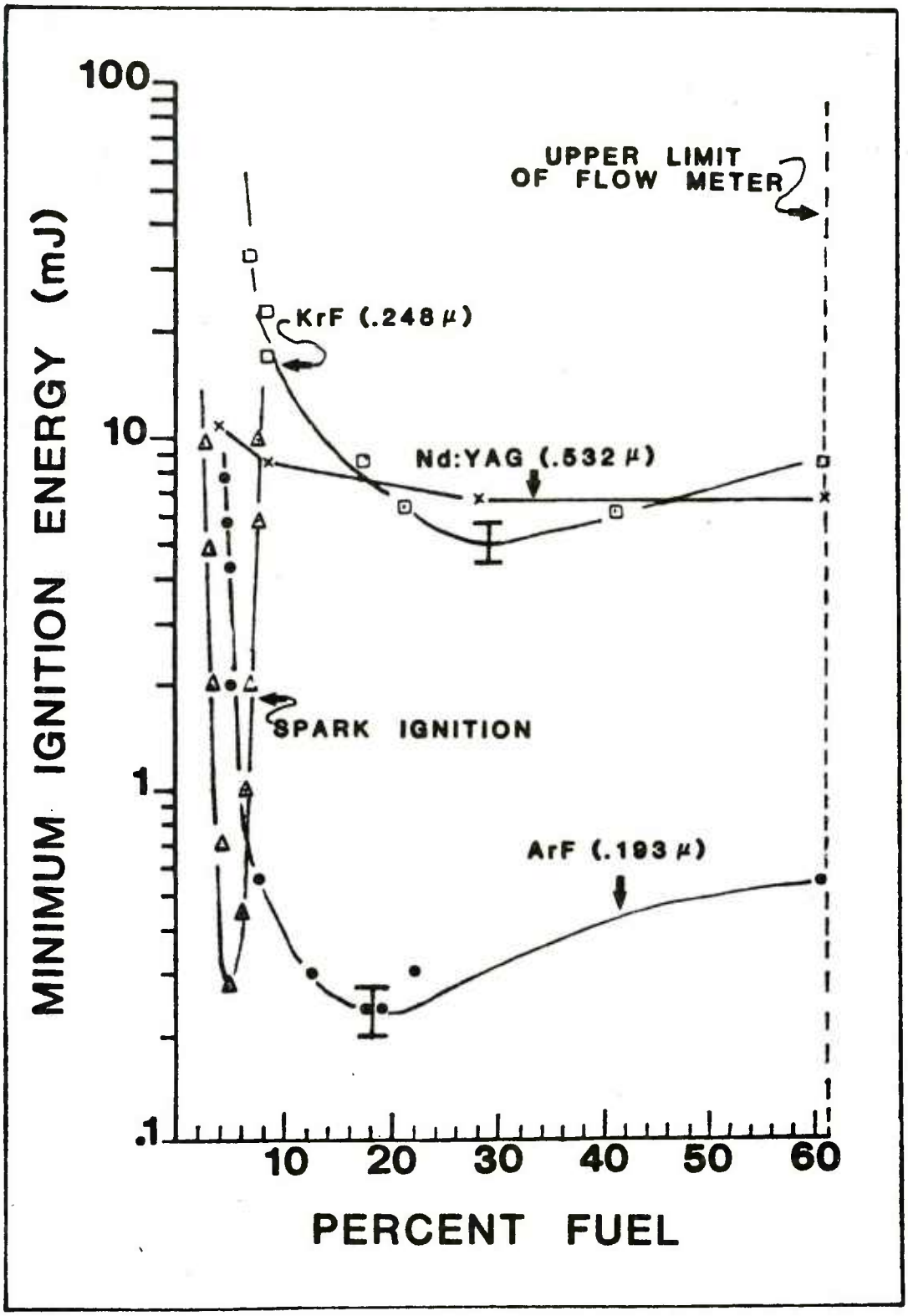

Figure 6. Dependence Of Minimum Ignition Energy On Percent Fuel For C_2H_2/Air. A Plot For C_3H_8/Air Spark Ignition In A Closed Bomb Is Included For Comparison.

either case, once the flame which is apparently beyond the flammability range is ignited, it burns as a diffusion flame rather than a pre-mixed flame. This apparent artifact, while yielding curves that are probably somewhat different in shape than they would be in a closed bomb experiment, does not affect substantially the point of this report, i.e., the demonstration of the existence and possible high efficiency of the ultraviolet laser multiphoton photochemical ignition source.

The results shown in Figure 6 further support attributing the excimer laser ignition mechanism to multiphoton photochemical effects since it is well known that C_2H_2 interacts significantly (much more than with either C_3H_8 or CH_4) with focussed ArF radiation by multiphoton absorption to give excited fragments like CH, C_2,[13] and C atoms.[12] To the extent that these experiments were done in a low-pressure photolysis apparatus and their detailed photochemical results may not directly apply and extend to the atmospheric pressure regime it should be noted that we have also observed emissions from excited CH, C_2, and C atoms both below and above ignition energy threshold while flowing a fuel-rich mixture during these experiments.

The Nd:YAG (532 nm) laser ignition mechanism appears to be due to gas breakdown/laser spark generation. Our observations are consistent with previous laser ignition studies using a Q-switched ruby laser[2,3] and a pulsed CO_2 laser[4] in that there was a very sharp laser energy threshold above which the spark size and blast wave would grow rapidly and ignition would always occur. Unfortunately, this phenomenon allows little control, especially with respect to the deposition of small amounts of energy into the focal volume. Close examination of Figures 2-6 reveals that, as noted before, there is only a small dependence of minimum ignition energy on the equivalence ratios and that all of the curves fall around the 9-10 mJ level, similar to the air breakdown value. Small variations in this energy are expected since gas breakdown is related to the ionization potential (I.P.) of the gases. Our results appear to be consistent since, for example, the C_2H_2/air mixture has a lower I.P. than does CH_4/N_2O and we find that the minimum ignition energies are around 7 mJ and 12 mJ for these two mixtures, respectively.

All of our results for laser ignition are summarized in Table 1, where the lowest minimum ignition energy for each of the plots given in Figures 2-6 is recorded. Due to the difficulty of smoothly varying the excimer laser energy, the uncertainty limits near the minimum points are somewhat large.

An important point to consider is that for the excimer ignition case, as mentioned before, only a relatively small fraction of the laser beam is absorbed (25% or less), while all of the spark ignition data is based on full absorption of available energy by the gases.[1] Thus, the photochemical ignition process is even more efficient than our data indicate, but it is difficult for us to quantify this since we cannot readily discern the difference between the fraction of laser energy absorbed and laser energy scattered. Since the multiphoton photochemical ignition is dependent on the

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[13] J.R. McDonald, A.P. Baranovski, and V.M. Donnely, "Multiphoton Vacuum Ultraviolet Laser Photodissociation of Acetylene: Emission From Electronically Excited Fragments," Chem. Phys., Vol. 33, p. 161, 1978.

TABLE 1. MINIMUM IGNITION ENERGIES FOR LASER IGNITION

Gas Mixture	Laser (λ)	Minimum Ignition Energy (mJ/pulse)
CH_4/air	ArF (193 nm)	>12
CH_4/N_2O	ArF (193 nm)	0.65
C_3H_8/air	ArF (193 nm)	4.8
C_3H_8/N_2O	ArF (193 nm)	0.91
C_2H_2/air	ArF (193 nm)	0.24
CH_4/air	KrF (248 nm)	>35
CH_4/N_2O	KrF (248 nm)	>35
C_3H_8/air	KrF (248 nm)	>35
C_3H_8/N_2O	KrF (248 nm)	16.0
C_2H_2/air	KrF (248 nm)	6.0
CH_4/air	Nd:YAG (532 nm)	9.5
CH_4/N_2O	Nd:YAG (532 nm)	12.0
C_3H_8/air	Nd:YAG (532 nm)	9.5
C_3H_8/N_2O	Nd:YAG (532 nm)	8.6
C_2H_2/air	Nd:YAG (532 nm)	7.0

power density at the focal volume, we expect that even lower laser energies will drive this process by using shorter focal length lenses and by using less divergent laser beams. Additionally, narrowing the broad-band (~0.6 nm) ArF laser to better match the multiphoton absorption profile, which is unknown at this time, should improve the efficiency of this process considerably. Another aspect of the need to focus the laser beams is that the interaction volume is well defined and could be a considerable distance from the optical access ports of a closed system. This may have profound practical implications, since now a photochemical ignition or combustion enhancement beam can be propagated through gas mixtures which are transparent to the beam, except in the focal point region where the multiphoton processes occur. Thus, the deleterious quenching effects due to nearby surfaces, like the windows, can be avoided.

An additional aspect to consider is that the relative efficiency of the photochemical ignition source as compared to electric spark or laser spark (breakdown) ignition may increase with decreasing pressure since the production of radicals and/or ions may be enhanced due to the decreasing effect of collisional quenching. Finally, it is clear that the comparison of our results with those for electric spark ignition is only approximate since our ignition criterion required that the energized gases stabilize on the slot burner. How much of an effect this is for the minimum ignition energy determination is not clear, but it can be argued that a more energetic ignition kernel is required to overcome the quenching effects of the burner.

To better understand this effect, photochemical ignition studies should be carried out in a closed bomb. In general, the prospects for significant increase in our understanding of the chemistry involved in ignition, especially photochemical ignition, appear to be excellent due to the substantial advances in optical diagnostic/laser techniques, and further experiments in this area are planned for the future.

IV. CONCLUSIONS

The following conclusions can be drawn from this work.

1. The two excimer lasers, ArF and KrF, as well as the Nd:YAG (second harmonic) laser can readily ignite most of the premixed fuel/oxidizer gas mixtures flowing through a slot burner when the laser beams are focused and have energies in the 0.2-40 mJ range.

2. Two distinct ignition mechanisms have been observed. The excimer lasers ignite the gas mixtures via a multiphoton photochemical pathway involving both fuel and oxidizer molecules and the amount of energy deposited into the reactive system can be well controlled. The Nd:YAG laser, however, causes gas breakdown with the formation of a substantial spark and blast wave even just at the threshold of breakdown. By the very nature of this process, it is impossible to deposit less energy into the system than what is absorbed at the breakdown threshold.

3. The ArF laser appears to be particularly efficient in igniting C_2H_2/air mixtures where the minimum ignition energy approaches 0.2 mJ and the primary photochemical processes involve the C_2H_2 molecule.

4. Since a substantial fraction of the laser beam is not absorbed during photochemically induced ignition, the process is probably even more efficient than our data indicate. Further improvements on efficiency should result from using shorter focal length lenses, a less divergent beam as well as by using a narrower linewidth excitation laser tuned to the multiphoton absorption transition which produces the required atoms, radicals, and/or ions that lead to the ignition of the gas mixture. The identification of these ignition precursors, as well as the search for the most photochemically efficient pathway to produce them, should be a most worthwhile endeavor. An enhancement in relative ignition efficiency may occur at lower pressures since collisional quenching processes will affect the production of radicals and/or ions to a lesser degree.

5. The focused nature of this process may have important, practical considerations since now the volume of photochemical activity is well-defined and can be a considerable distance from quenching surfaces. Also, depending on the laser wavelengths and chemical species, the laser beam can be transmitted undisturbed due to the lack of one-photon interactions until the focal point region is reached where the multiphoton processes are induced.

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